

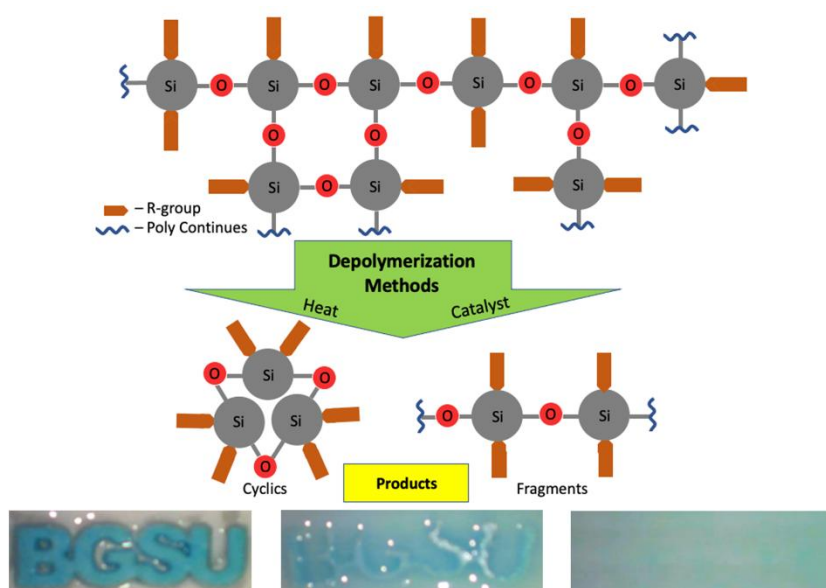


Towards a Circular Economy in Silicones through the Depolymerization and Repolymerization of Siloxanes, Alkoxysilanes, and Dynamic Bonding Motifs

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This presentation will showcase our recent work on the development of circular methodologies for siloxane-based elastomers and resins to demonstrate the potential industrial feasibility of silicone recycling/upcycling. We have developed an efficient methodology to depolymerize polysiloxanes of various forms and cross-link densities to cyclic siloxanes of D₄, D₅, and D₆ with an emphasis towards D₄ development and then demonstrate their repolymerization back to polysiloxane structures. The depolymerization processes are conducted using a cocktail of fluoride ions from tetrabutylammonium fluoride or from photoreleasable fluorides from phenacyl photocages in THF or MEK and other intermediate high swell solvents. The reactions are highly efficient with conversion to cyclomers taking place in as little as 30 min through an equilibration process, with quenching by CaCl₂ to remove active fluoride ions and lock the cyclic structures. The conversion to cyclomers are verified by ²⁹Si NMR and GCMS spectroscopic techniques. Traditional methods such as triflic acid initiated polymerization have shown effective repolymerization to polysiloxanes containing M_w up to 8600 Da. We also demonstrate methods for the mechanochemical transformation of siloxane polymers to cyclic compounds and also the conversion of siloxanes directly to alkoxysilanes via catalytic processes. Lastly, we will show how the incorporation of light responsive cross-linkers into siloxane systems can enable dynamic re-processible polymeric systems by losing and then restoring mechanical properties for adhesives and self-healable materials.



References

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- [2] B. Rupasinghe, J. C. Furgal, *Polym. Int.* **2022**, 71, 521–531.